Heat conductivity of the spin-Peierls compounds TiOCl and TiOBr

Hlubek, N.; Sing, M.; Glawion, S.; Claessen, R.; van Smaalen, S.; van Loosdrecht, P. H. M.; Buechner, B.; Hess, C.; Büchner, B.

Published in:
Physical Review. B: Condensed Matter and Materials Physics

DOI:
10.1103/PhysRevB.81.144428

Publication date:
2010

Heat conductivity of the spin-Peierls compounds TiOCl and TiOBr

N. Hlubek,[1] M. Sing,[2] S. Glawion,[2] R. Claessen,[2] S. van Smaalen,[3] P. H. M. van Loosdrecht,[4] B. Büchner,[1] and C. Hess[1]
[1]*IFW-Dresden, Institute for Solid State Research, P.O. Box 270116, D-01171 Dresden, Germany*
[2]*Experimentelle Physik 4, Universität Würzburg, 97074 Würzburg, Germany*
[3]*Laboratory of Crystallography, Universität Bayreuth, 95440 Bayreuth, Germany*
[4]*Zernike Institute for Advanced Materials, University of Groningen, 9747 AG Groningen, The Netherlands*
(Received 10 February 2010; published 28 April 2010)

We report experimental results on the heat conductivity κ of the $S=1/2$ spin chain compounds TiOBr and TiOCl for temperatures 5 K$<T<$300 K and magnetic fields up to 14 T. Surprisingly, we find no evidence of a significant magnetic contribution to κ, which is in stark contrast to recent results on $S=1/2$ spin chain cuprates. Despite this unexpected result, the thus predominantly phononic heat conductivity of these spin-Peierls compounds exhibits a very unusual behavior. In particular, we observe strong anomalies at the phase transitions T_{c1} and T_{c2}. Moreover, we find an overall but anisotropic suppression of κ in the intermediate phase which extends even to temperatures higher than T_{c2}. An external magnetic field causes a slight downshift of the transition at T_{c1} and enhances the suppression of κ up to T_{c2}. We interpret our findings in terms of strong spin-phonon coupling and phonon scattering arising from spin-driven lattice distortions.

DOI: 10.1103/PhysRevB.81.144428 PACS number(s): 66.70.−f, 75.40.Gb, 75.10.Pq, 68.65.−k

I. INTRODUCTION

Understanding low-dimensional quantum spin-1/2 systems is one of the challenges of contemporary condensed matter physics. In particular, transition metal oxides provide a rich playground for studying novel phenomena, arising from the interplay between lattice, orbital, spin, and charge degrees of freedom. The recent discovery of a substantial magnetic heat conductivity κ_{mag} in one-dimensional (1D) quantum spin systems,[1–11] together with the theoretical prediction of ballistic transport in 1D $S=1/2$ Heisenberg chains,[12–14] has caused intense experimental and theoretical research on the behavior of these systems. The best experimental realizations of $S=1/2$ systems showing magnetic heat transport are up to now found among copper oxides (cuprates) such as the spin chains $SrCuO_2$ and Sr_2CuO_3 (Refs. 2 and 11) and the spin ladder compounds $(Ca,La,Sr)_{14}Cu_{24}O_{41}$.[1,3,5,8] Characteristic for the cuprate chain systems is a Cu $3d^9$ configuration which gives rise to $S=1/2$ and a large exchange coupling $J/k_B \approx 2000$ K along the chain/ladder direction. As a consequence of this quasi-1D magnetic structure, these systems exhibit a strongly anisotropic thermal transport behavior. Perpendicular to the low-dimensional spin structure a typical phononic heat conductivity κ_{ph} is found. However, parallel to the low-dimensional spin structure the heat conductivity is strongly enhanced even up to room temperature since a large κ_{mag} adds to κ_{ph}. These in many aspects excellent realizations of $S=1/2$ Heisenberg chains do not undergo a spin-Peierls transition, i.e., a transition to a spin-dimerized ground state at the expense of a lattice distortion that normally should arise from the spin-phonon coupling of a spin chain and the phonon system in which it is embedded. Surprisingly, only one Cu-based spin system, $CuGeO_3$, is known to exhibit a spin-Peierls transition.[15] The exchange energy of this compound is $J/k_B \approx 160$ K,[16] and the transition to the nonmagnetic ground state is at $T_c \approx 14$ K. The heat conductivity of $CuGeO_3$ has been studied by several groups[17–19] with controversial results. 1D magnetic heat conductivity has been suggested to give rise to a significantly enhanced heat conductivity at $T<T_c$.[17] However, the observed low-temperature peak has been shown to be present both in the heat conductivity parallel and perpendicular to the chain and thus can be rationalized in terms of phononic transport alone.[19]

Also spin $S=1/2$ systems, but based on early transition metal ions with electronic configuration $3d^1$, the titanium oxyhalides TiOX, with X=Br or Cl shifted recently into focus. These compounds are considered as good realizations of $S=1/2$ spin chains, which are formed by direct overlap of Ti t_{2g} orbitals along the crystallographic b direction[20–22] with rather high magnetic exchange coupling $J(\mathrm{Cl}) \approx 676$ K (Refs. 20 and 21) and $J(\mathrm{Br}) \approx 375$ K.[23–25] The compounds undergo two phase transitions T_{c1}, T_{c2},[23,26] where the lower one, at T_{c1}, leads to a nonmagnetic dimerized state,[20] which is accompanied by a doubling of the unit cell.[24,27] These features thus render the Ti oxyhalides the second (besides $CuGeO_3$) type of inorganic compounds which undergoes a spin-Peierls transition. However, as compared to $CuGeO_3$ the dimerized state occurs at much higher temperatures, viz. $T_{c1,\mathrm{Cl}}=67$ K for TiOCl and $T_{c1,\mathrm{Br}}=28$ K for TiOBr. However, several experimental results are inconsistent with a canonical spin-Peierls scenario. There are two successive phase transitions and the transition to the nonmagnetic state at T_{c1} is of first[24,26,27] and not of second order as in $CuGeO_3$. Interestingly, in the intermediate regime between T_{c1} and T_{c2} an incommensurate superstructure is found.[28–31] Above T_{c2} ($T_{c2,\mathrm{Cl}}=91$ K for TiOCl and $T_{c2,\mathrm{Br}}=48$ K for TiOBr), the system is in a pseudo-spin-gap regime up to a characteristic temperature T^* which for TiOCl extends up to $T^* \approx 135$ K with a large singlet-triplet energy gap of $E_g=430$ K.[30,32–34] First explanations of the intermediate phase proposed orbital fluctuations, but this has been ruled out by optical measurements in combination with cluster calculations that showed that the crystal field splitting is large enough to quench the orbital degree of freedom.[23] Recent explanations focus on the interplay between intrachain and interchain frustrations and a related dimensionality crossover.[28,30,35–38]

The relatively high magnetic exchange constants of the titanium oxyhalides render them good noncuprate candidates for exhibiting a sizeable magnetic heat conductivity arising

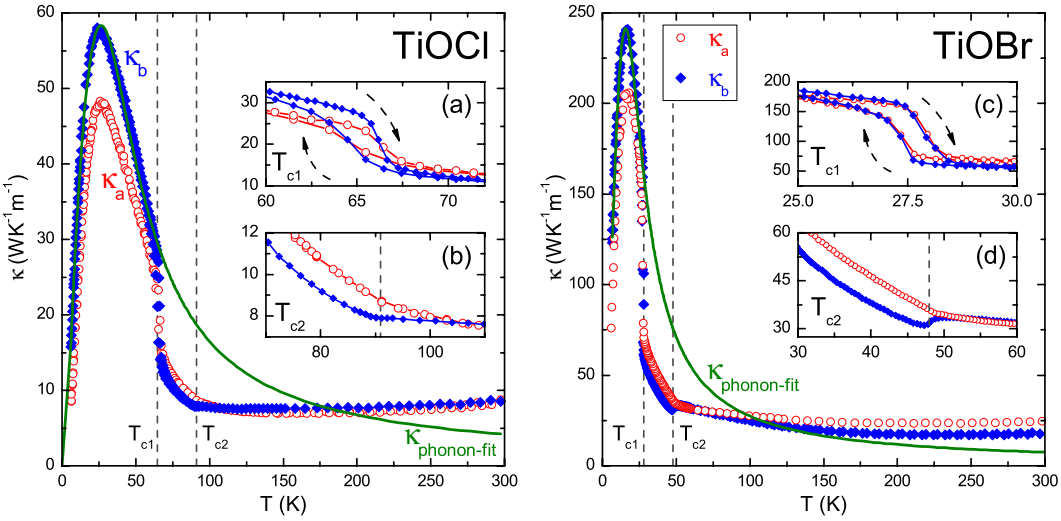

FIG. 1. (Color online) Thermal conductivities κ_a (○) and κ_b (◆) of TiOCl and TiOBr as a function of T. The insets depict the behavior of the thermal conductivity around the respective phase transitions. Insets (a) and (c) show the hysteretic behavior around the phase transition at T_{c1}, which is characteristic for a first-order transition. The arrows mark the corresponding curves for cooling and heating. The lower insets (b) and (d) show κ around the transition at T_{c2} also measured for cooling and heating. It is without hysteresis and therefore the transition is of second order.

from the 1D $S=1/2$ spin chains. In this paper, we experimentally investigate the thermal conductivity κ of TiOCl and TiOBr with a special focus on potentially arising magnetic contributions to κ. Surprisingly, no indication for magnetic heat transport is observed and we find instead that κ is dominated by phononic heat conduction. However, strong anomalies occur at the phase transitions T_{c1} and T_{c2} and we find an overall suppression of the phononic κ, which is anisotropic in the incommensurate phase and which extends to temperatures higher than T_{c2}. For TiOBr the application of an external magnetic field of 14 T slightly shifts T_{c1} toward lower temperature and causes a weak further suppression of κ in the intermediate regime.

II. EXPERIMENT

Single crystals of TiOCl and TiOBr were synthesized by a chemical vapor transport technique leading to small platelike crystals.[39] The crystallinity was checked by x-ray diffraction. Typical crystal dimensions are a few mm^2 in the ab plane but only around 20 μm along the c axis. Rectangular samples with typical dimensions of $(2\times1\times0.02)$ mm^3 with the longest side being parallel to the a and b axis, respectively, were cut from the crystal plates. Measurements of the thermal conductivity as a function of temperature T in the range of 7–300 K were performed with a standard four-probe technique.[40] Because of the small thickness of the crystals the usual uncertainty of 10% for κ due to the error in the determination of the crystal geometry is exceeded by some extent. Furthermore, the small thickness along the c axis also prevented to measure κ along this direction. In order to compare the anisotropy of κ along the a and b directions, the individual samples were cut from the same crystal plate, thus keeping the relative error between the two directions small. The mounting of TiOBr into the heat conductivity probe was performed under Argon atmosphere in order to minimize degradation of the sample.

III. RESULTS

Figure 1 shows the temperature dependence of the thermal conductivities along the a and b axes (κ_a and κ_b) of TiOCl and TiOBr in zero magnetic field. We focus first on the results for TiOCl which are shown in the left panel of Fig. 1. A first glance at the data already suggests that the temperature dependence of κ is governed by the two phase transitions at T_{c1} and T_{c2}, which divide the data into three regimes. At low temperature the heat conductivity parallel to the chains, κ_b, exhibits a strong peak at $\sim$25 K with a maximum value $\kappa_b \approx 58$ Wm^{-1} K^{-1} which is a typical feature of a phononic heat conductivity κ_{ph} at low temperature. It arises from two competing effects: At very low temperature the mean free path of phonons is determined by the crystal boundaries and defects and therefore is practically T independent. Hence, κ_{ph} increases due to the increasing number of phonons. At higher temperature the mean free path is T dependent as the number of umklapp processes rises exponentially. This overcompensates the effect of a rising phonon population and thus κ_{ph} decreases with further rising T, i.e., κ_{ph} usually shows a maximum. Interestingly, κ_b deviates from this conventional behavior at T_{c1}, where a sharp drop occurs to about 60% of the value of κ_b at just below the transition. In the intermediate phase κ_b continuously decreases further with rising T just until T_{c2} is reached. Upon rising T through T_{c2} we find that κ_b changes slope and exhibits a weak increase in the entire high temperature phase, i.e., at $T>T_{c2}$, up to room temperature.

A very similar temperature dependence is observed in the heat conductivity perpendicular to the chains, κ_a. In this case, the peak at $T\approx25$ K is somewhat smaller ($\kappa_{a,\max}$

≈ 48 $\mathrm{Wm^{-1}\,K^{-1}}$) than that in κ_b. A similarly sharp drop as in the latter occurs at T_{c1}. However, the actual drop at the transition is relatively weaker as in the other direction. Interestingly, despite a similar further decrease of κ_a when rising T toward T_{c2} as in κ_b, we find that κ_a remains always somewhat *larger* in this intermediate regime. The slope of κ_a changes at T_{c2}, but remains negative up to $T \approx 150$ K, in contrast to the findings for κ_b [cf. Fig. 1(b)].

Before discussing these peculiarities in detail, we briefly summarize the results for TiOBr which are shown in the right panel of Fig. 1. The general T dependence of κ has large similarities with that of TiOCl, including the observed anomalies. There are, however, slight differences which are worth to be pointed out: First, the phononic peak of both κ_a and κ_b of TiOBr is by a factor of about 4 larger than that in TiOCl and is located at somewhat lower temperature (~ 17 K). Both features point to a lower defect density in the case of TiOBr. This is corroborated by room temperature x-ray diffraction, which showed much sharper spots for TiOBr. Second, at $T < T_{c2}$ the anisotropy between κ_a and κ_b is similar to that of TiOCl. More specifically, at $T < T_{c1}$ we find $\kappa_a < \kappa_b$, and $\kappa_a > \kappa_b$ at $T_{c1} < T < T_{c2}$, i.e., the drop at T_{c1} and the reduction of κ are relatively stronger in κ_b than that in κ_a. Interestingly, the anomaly in κ_b at T_{c2} is much stronger than that in TiOCl since a clear dip is observable at the transition [cf. Fig. 1(d)]. Moreover, in contrast to TiOCl we observe that both κ_a and κ_b decrease with rising temperature at $T > T_{c2}$ up to room temperature where κ_a remains slightly larger than κ_b.

IV. DISCUSSION

The overall very weak anisotropy of the κ data suggests without further analysis the unexpected conclusion that magnetic heat transport in the spin chains of this material is negligible in both TiOCl and TiOBr. Otherwise a significant enhancement of κ_b with respect to κ_a should occur since heat transport by magnetic excitations is only expected along the 1D spin chain, i.e., parallel to b. One might speculate that the weak anisotropy that is present in the low-temperature regime $T < T_{c1}$ is the indication of a weak magnetic contribution along b, which could give rise to the observed $\kappa_b > \kappa_a$. However, the observed anisotropy by a factor ~ 1.2 matches that of other phononic heat conductors[40,41] and can conventionally be explained by differences in the phonon velocity.

At higher temperatures ($T > T_{c1}$) magnetic contributions appear even more unlikely, since in all cases $\kappa_b \lesssim \kappa_a$. However, in this regime a small magnetic contribution to κ_b might still be present if the expected anisotropy was masked by differences in the phononic transport along the two crystallographic directions. Concentrating only on the thermal conductivity κ_b we estimate the thus maximum possible κ_{mag} by performing a phononic fit based on the so-called Callaway model[42] to the low-temperature peak and extrapolate this fit toward room temperature.[43] The fit is depicted by the solid line in Fig. 1 and yields a very good agreement up to T_{c1} but deviates strongly from the data at higher temperatures. In particular, at high temperatures ($T \gtrsim 180$ K) the fit is clearly lower than the data. We use the difference between the fit $\kappa_{\mathrm{ph,Fit}}$ and the data at room temperature to obtain an upper estimate for the possible magnetic contributions $\kappa_{\mathrm{mag}} = \kappa_b - \kappa_{\mathrm{ph,Fit}}$. In order to analyze the thermal transport we estimate the magnetic mean free path l_{mag} using an approximation of κ_{mag} of a $S=1/2$ Heisenberg chain,[2,10]

$$\kappa_{\mathrm{mag}} = \frac{2 n_s k_B^2}{\pi \hbar} l_{\mathrm{mag}} T \int_0^{J\pi/2k_BT} x^2 \frac{\exp(x)}{[\exp(x)+1]^2} dx, \qquad (1)$$

where n_s is a geometrical factor that counts the number of chains per unit area. For both compounds this yields a negligibly small mean free path of only two to three lattice constants.[54] Considering the fact that the Callaway model usually underestimates κ_{ph} at room temperature[44,45] and that $\kappa_b \lesssim \kappa_a$ at higher temperature, any realistic value for the mean free path should be even smaller which essentially rules out magnetic transport in the Ti oxyhalides.

There are not many scenarios which straightforwardly explain this unexpected result. The absence of magnetic heat conduction in magnetic materials has been discussed by Sanders and Walton in terms of a very large magnon-phonon relaxation time.[46] It is obvious that this situation cannot be realized in Ti oxyhalides since a significant spin-phonon coupling must be present in these compounds to allow for a spin-Peierls transition at considerably high temperatures. In fact, it is therefore more reasonable to explain the absence of magnetic heat conduction by a particularly strong spin-phonon coupling, which gives rise to strong scattering of spin excitations and thus prevents the magnetic heat conduction. One might speculate that even more exotic excitations such as orbital fluctuations are relevant for suppressing κ_{mag}. We point out, however, that orbital excitations have been shown to be unimportant for the low-energy physics in these compounds.[22,23]

The negligible magnetic heat conduction in the Ti oxyhalides implies that the unusual temperature dependence and also the slight anisotropy should be rationalized in terms of pure phonon heat conduction, which has been proven to be a sensitive probe to peculiarities of the lattice such as superstructures and disorder.[40,41,47,48] The considerable jump in κ at T_{c1} clearly indicates that the phonon heat conduction in the intermediate phase is strongly suppressed with respect to that of the commensurate dimerized phase at $T < T_{c1}$ where ordinary phonon heat conduction is observed. This reflects the abrupt transition toward a lattice with strongly disturbed periodicity and anharmonicity, which causes enhanced phonon scattering and is entirely consistent with the incommensurate lattice distortion in this regime.[29,32,49] We have investigated the nature of this phase transition at T_{c1} and find for both compounds a clear hysteretic behavior, which confirms the transition being of first order [see Figs. 1(a) and 1(c)]. Such first-order character has already been reported from magnetic susceptibility, specific heat, thermal expansion, and x-ray data of the superstructure satellites.[22,23,26,27] Since the magnetic exchange is smallest in TiOBr we have searched for possible effects of a magnetic field on κ_b. As is depicted in Fig. 2 a magnetic field of $B=14$ T along the b direction has only little influence on the thermal conductivity $\kappa_{b,14\ \mathrm{T}}$. However, we detect a slight downshift of the phase transition

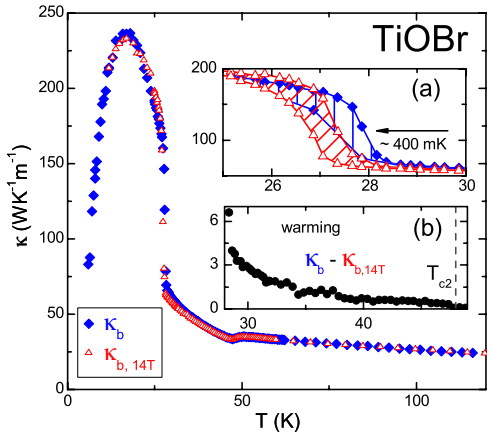

FIG. 2. (Color online) Thermal conductivity κ_b as a function of T in TiOBr with $[\kappa_{b,14\ \mathrm{T}}\ (\triangle)]$ and without $[\kappa_b\ (\blacklozenge)]$ an applied magnetic field of 14 T along the chain direction. Inset (a) illustrates the shift of T_{c1} toward lower temperatures in the presence of a magnetic field. In inset (b) $\Delta\kappa=\kappa_b-\kappa_{b,14\ \mathrm{T}}$ shows the decreasing influence of the magnetic field on κ_b in the intermediate regime. The curves used in the subtraction are from the measurements that approach the phase transitions from low temperatures.

at T_{c1} by ~400 mK, which is consistent with a downshift of ~130 mK that has been reported from x-ray diffraction at $B=10$ T for TiOCl.[50] Moreover, starting at T_{c1}, $\kappa_{b,14\ \mathrm{T}}$ is slightly smaller compared to the measurement without field, but gradually approaches it for increasing temperature. In Fig. 2(b) the difference $\Delta\kappa=\kappa_b-\kappa_{b,14\ \mathrm{T}}$ between both curves is shown, illustrating the decreasing influence of the magnetic field, until it vanishes at T_{c2}. This suggests that the spin-induced incommensurate lattice distortion in this intermediate phase is further enhanced by an external magnetic field.

The thermal conductivity across the phase transitions at T_{c2} shown in more detail in Figs. 1(b) and 1(d), does not exhibit a hysteretic behavior which is indicative of a second-order transition. The overall impact of this transition on κ is much smaller than that at T_{c1}. Interestingly, in the high-temperature phase above T_{c2} the thermal conductivity appears still significantly suppressed with respect to the low-temperature phase at $T<T_{c1}$. In Fig. 1 this is clearly seen when comparing the data to the phononic fit, which remains much larger than κ up to $T^*\sim 100$ K and $T^*\sim 180$ K for TiOBr and TiOCl, respectively. Only at higher temperatures a more typical behavior is observed with $\kappa_{\mathrm{ph,Fit}}<\kappa$. The apparent suppression of κ in the regime $T_{c2}\lesssim T\lesssim T^*$ clearly indicates that strong phonon scattering occurs despite the absence of any static long-range lattice distortions. A reasonable origin of this enhanced scattering is precursors of the spin-Peierls transition, either as short-range static lattice distortions or as slowly fluctuating precursors (soft phonon type). This is consistent with the pseudogap seen in magnetic resonance measurements[21,32,33] and incommensurate structural fluctuations found by x-ray diffraction.[49]

In both compounds the suppression of κ in the intermediate phase is clearly anisotropic, since the drop of κ_b at T_{c1} is relatively stronger as compared to κ_a and $\kappa_b<\kappa_a$ in the entire phase, where κ_b of TiOBr even shows a local minimum at T_{c2}. Similar anisotropic scattering has previously been observed, e.g., in stripe-ordering compounds which possess anisotropic correlation lengths of the stripe order close to the transition.[41] The stronger suppression of κ_b than κ_a in the present case can be understood by looking at the modulation amplitudes for TiOCl (Ref. 49) and TiOBr in the incommensurate phase.[29] Those indicate that the shifts of the atoms out of the periodic position of the structure at room temperature are larger in the direction of the b axis than those along the a axis. The resulting larger anharmonicity along b is likely causing increased scattering and therefore the observed lower thermal conductivity.

There is a slight difference in the thermal conductivity between both compounds near room temperature where phonon scattering arising from the spin-Peierls transition can be considered to be relatively weak. For TiOBr the slope of κ is negative while it is positive for TiOCl. At the same time the absolute value of κ is significantly higher in TiOBr. This corroborates the previous conclusion that our TiOBr crystals have a lower defect density than the TiOCl ones because the observed temperature dependence of κ for TiOBr is much closer to the expected $\propto T^{-1}$ decrease of a clean phonon heat conductor.[51] On the other hand, the lower κ of TiOCl with a weak positive slope is typical for more disordered heat conductors, where also rather small contributions to κ, such as heat transport by optical phonons[52] become relevant.

V. SUMMARY

In conclusion, we have shown that the magnetic thermal conductivity in the TiOX is negligible due to strong spin-phonon scattering. The heat transport can thus be understood in terms of pure phononic conductivity. At the phase transitions we find strong anomalies which are consistent with the lattice distortions. Starting at low temperatures, the first phase transition T_{c1} toward the dimerized state can be shifted to lower temperatures by an external magnetic field. Additionally, this leads to a slight suppression of the thermal conductivity throughout the intermediate regime and gradually gets smaller when approaching T_{c2}. Comparing the measurements along the different crystallographic directions in this regime, the stronger suppression along κ_b for both compounds is consistent with a higher incommensurability of the lattice in this direction. Finally, by a comparison of the extrapolated thermal conductivity from a phononic model to the measurement at higher temperatures it was argued that the thermal conductivity is still suppressed up to a temperature T^* which is either a sign of short-range lattice distortions or phonon softening.

ACKNOWLEDGMENTS

We thank Daniel Khomskii and Daniele Fausti for valuable discussions. This work was supported by the Deutsche Forschungsgemeinschaft through Grants No. HE3439/7, No. SM55/15, and No. CL124/6, through the Forschergruppe FOR912 (Grant No. HE3439/8), and by the European Commission through the NOVMAG project (Grant No. FP6-032980).

[1] A. V. Sologubenko, K. Giannò, H. R. Ott, U. Ammerahl, and A. Revcolevschi, Phys. Rev. Lett. **84**, 2714 (2000).
[2] A. V. Sologubenko, K. Giannò, H. R. Ott, A. Vietkine, and A. Revcolevschi, Phys. Rev. B **64**, 054412 (2001).
[3] C. Hess, C. Baumann, U. Ammerahl, B. Büchner, F. Heidrich-Meisner, W. Brenig, and A. Revcolevschi, Phys. Rev. B **64**, 184305 (2001).
[4] C. Hess, U. Ammerahl, C. Baumann, B. Büchner, and A. Revcolevschi, Physica B **312-313**, 612 (2002).
[5] C. Hess, H. ElHaes, B. Büchner, U. Ammerahl, M. Hücker, and A. Revcolevschi, Phys. Rev. Lett. **93**, 027005 (2004).
[6] C. Hess, C. Baumann, and B. Büchner, J. Magn. Magn. Mater. **290-291**, 322 (2005).
[7] P. Ribeiro, C. Hess, P. Reutler, G. Roth, and B. Büchner, J. Magn. Magn. Mater. **290-291**, 334 (2005).
[8] C. Hess, P. Ribeiro, B. Büchner, H. ElHaes, G. Roth, U. Ammerahl, and A. Revcolevschi, Phys. Rev. B **73**, 104407 (2006).
[9] C. Hess, H. ElHaes, A. Waske, B. Büchner, C. Sekar, G. Krabbes, F. Heidrich-Meisner, and W. Brenig, Phys. Rev. Lett. **98**, 027201 (2007).
[10] C. Hess, Eur. Phys. J. Spec. Top. **151**, 73 (2007).
[11] N. Hlubek, P. Ribeiro, R. Saint-Martin, A. Revcolevschi, G. Roth, G. Behr, B. Büchner, and C. Hess, Phys. Rev. B **81**, 020405(R) (2010).
[12] X. Zotos, F. Naef, and P. Prelovsek, Phys. Rev. B **55**, 11029 (1997).
[13] A. Klümper and K. Sakai, J. Phys. A **35**, 2173 (2002).
[14] F. Heidrich-Meisner, A. Honecker, D. C. Cabra, and W. Brenig, Phys. Rev. B **68**, 134436 (2003).
[15] M. Hase, I. Terasaki, and K. Uchinokura, Phys. Rev. Lett. **70**, 3651 (1993).
[16] K. Fabricius, A. Klümper, U. Löw, B. Büchner, T. Lorenz, G. Dhaleene, and A. Revcolevschi, Phys. Rev. B **57**, 1102 (1998).
[17] Y. Ando, J. Takeya, D. L. Sisson, S. G. Doettinger, I. Tanaka, R. S. Feigelson, and A. Kapitulnik, Phys. Rev. B **58**, R2913 (1998).
[18] A. M. Vasil'ev, M. I. Kaganov, V. V. Pryadun, G. Dhalenne, and A. Revcolevschi, JETP Lett. **66**, 868 (1997).
[19] M. Hofmann, T. Lorenz, A. Freimuth, G. Uhrig, H. Kageyama, Y. Ueda, G. Dhalenne, and A. Revcolevschi, Physica B **312-313**, 597 (2002).
[20] A. Seidel, C. A. Marianetti, F. C. Chou, G. Ceder, and P. A. Lee, Phys. Rev. B **67**, 020405(R) (2003).
[21] V. Kataev, J. Baier, A. Möller, L. Jongen, G. Meyer, and A. Freimuth, Phys. Rev. B **68**, 140405(R) (2003).
[22] M. Hoinkis *et al.*, Phys. Rev. B **72**, 125127 (2005).
[23] R. Rückamp, J. Baier, M. Kriener, M. W. Haverkort, T. Lorenz, G. S. Uhrig, L. Jongen, A. Möller, G. Meyer, and M. Grüninger, Phys. Rev. Lett. **95**, 097203 (2005).
[24] T. Sasaki, M. Mizumaki, K. Kato, Y. Watabe, Y. Nishihata, M. Takata, and J. Akimitsu, J. Phys. Soc. Jpn. **74**, 2185 (2005).
[25] C. Kato, Y. Kobayashi, and M. Sato, J. Phys. Soc. Jpn. **74**, 473 (2005).
[26] J. Hemberger, M. Hoinkis, M. Klemm, M. Sing, R. Claessen, S. Horn, and A. Loidl, Phys. Rev. B **72**, 012420 (2005).
[27] M. Shaz, S. van Smaalen, L. Palatinus, M. Hoinkis, M. Klemm, S. Horn, and R. Claessen, Phys. Rev. B **71**, 100405(R) (2005).
[28] E. T. Abel, K. Matan, F. C. Chou, E. D. Isaacs, D. E. Moncton, H. Sinn, A. Alatas, and Y. S. Lee, Phys. Rev. B **76**, 214304 (2007).
[29] S. van Smaalen, L. Palatinus, and A. Schönleber, Phys. Rev. B **72**, 020105(R) (2005).
[30] J. P. Clancy, B. D. Gaulin, K. C. Rule, J. P. Castellan, and F. C. Chou, Phys. Rev. B **75**, 100401(R) (2007).
[31] J. P. Clancy, B. D. Gaulin, and F. C. Chou, Phys. Rev. B **81**, 024411 (2010).
[32] T. Imai and F. Chou, arXiv:cond-mat/0301425 (unpublished).
[33] S. R. Saha, S. Golin, T. Imai, and F. C. Chou, J. Phys. Chem. Solids **68**, 2044 (2007).
[34] P. J. Baker *et al.*, Phys. Rev. B **75**, 094404 (2007).
[35] D. Fausti *et al.*, Phys. Rev. B **75**, 245114 (2007).
[36] R. Macovez, J. Luzon, J. Schiessling, A. Sadoc, L. Kjeldgaard, S. van Smaalen, D. Fausti, P. H. M. van Loosdrecht, R. Broer, and P. Rudolf, Phys. Rev. B **76**, 205111 (2007).
[37] D. Mastrogiuseppe and A. Dobry, Phys. Rev. B **79**, 134430 (2009).
[38] Y.-Z. Zhang, H. O. Jeschke, and R. Valentí, Phys. Rev. Lett. **101**, 136406 (2008).
[39] H. Schaefer, F. Wartenpfuhl, and E. Weise, Z. Anorg. Allg. Chem. **295**, 268 (1958).
[40] C. Hess, B. Büchner, U. Ammerahl, and A. Revcolevschi, Phys. Rev. B **68**, 184517 (2003).
[41] C. Hess, B. Büchner, M. Hücker, R. Gross, and S.-W. Cheong, Phys. Rev. B **59**, R10397 (1999).
[42] J. Callaway, Phys. Rev. **113**, 1046 (1959).
[43] The fit was done for $T<T_{c1}$.
[44] R. Berman and J. Brock, Proc. R. Soc. London, Ser. A **289**, 46 (1965).
[45] M. Asen-Palmer, K. Bartkowski, E. Gmelin, M. Cardona, A. P. Zhernov, A. V. Inyushkin, A. Taldenkov, V. I. Ozhogin, K. M. Itoh, and E. E. Haller, Phys. Rev. B **56**, 9431 (1997).
[46] D. J. Sanders and D. Walton, Phys. Rev. B **15**, 1489 (1977).
[47] K. Berggold, M. Kriener, P. Becker, M. Benomar, M. Reuther, C. Zobel, and T. Lorenz, Phys. Rev. B **78**, 134402 (2008).
[48] D. Cassel, C. Hess, B. Büchner, M. Hücker, R. Gross, O. Friedt, and S.-W. Cheong, J. Low Temp. Phys. **117**, 1083 (1999).
[49] A. Schönleber, S. van Smaalen, and L. Palatinus, Phys. Rev. B **73**, 214410 (2006).
[50] A. Krimmel *et al.*, Phys. Rev. B **73**, 172413 (2006).
[51] R. Berman, *Thermal Conduction in Solids* (Clarendon, Oxford, 1976).
[52] C. Hess and B. Büchner, Eur. Phys. J. B **38**, 37 (2004).
[53] A. V. Sologubenko, H. R. Ott, G. Dhalenne, and A. Revcolevschi, Europhys. Lett. **62**, 540 (2003).
[54] A different approach to estimate κ_{mag} that follows an analysis described in Ref. 53 using the thermal Drude weight (Ref. 13) yields the same result.